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Non-Markovian decay of a three-level cascade atom in a structured reservoir

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The dynamics of a three-level atom in a cascade (or ladder) configuration with both transitions coupled to a single structured reservoir of quantized electromagnetic field modes is treated using Laplace transform methods applied to the coupled amplitude equations. In this system two-photon excitation of the reservoir occurs, and both sequences for emitting the two photons are allowed and included in the theory. An integral equation is found to govern the complex amplitudes of interest. It is shown that the dynamics of the atomic system is completely determined in terms of reservoir structure functions, which are products of the mode density with the coupling constant squared. This dependence on reservoir structure functions rather than on the mode density or coupling constants alone, shows that it may be possible to extend pseudomode theory to treat multiphoton excitation of a structured reservoir—pseudomodes being introduced in one-one correspondence with the poles of reservoir structure functions in the complex frequency plane. A general numerical method for solving the integral equations based on discretizing frequency space, and applicable to different structured reservoirs such as high-Q cavities and photonic band-gap systems, is presented. An application to a high-Q-cavity case with identical Lorentzian reservoir structure functions is made, and the non-Markovian decay of the excited state shown. A formal solution to the integral equations in terms of right and left eigenfunctions of a non-Hermitian kernel is also given. The dynamics of the cascade atom, with the two transitions coupled to two separate structured reservoirs of quantized electromagnetic field modes, is treated similarly to the single structured reservoir situation. Again the dynamics only depends on reservoir structure functions. As only one sequence of emitting the two photons now occurs, the integral equation for the amplitudes can be solved analytically. The non-Markovian decay of the excited state is shown for the same high-Q-cavity case of identical Lorentzian reservoir structure functions, and differs from that for the single reservoir situation.

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I. INTRODUCTION

The quantum optical behavior of atomic systems coupled to a continuum of the quantized electromagnetic field modes has been studied since the early days of quantum physics. The quantum electromagnetic field is a large system, which can be treated as a bath or reservoir. In most cases the atom-field coupling constants and the electromagnetic field mode density are slowly varying functions of frequency, and the dynamics of the atomic system can be treated via Markovian master equations [1,2] or equivalent methods such as quantum Langevin equations (see, e.g., Ref. [2] for details of these standard methods). These techniques are based on quantum electromagnetic field states with no special distinction for any particular mode in terms of photon occupation number, such as thermal states or broad-band squeezed states. Naturally, if one mode of the electromagnetic field was in a special state, such as a large amplitude coherent state (as in the case where the atom is also coupled to a laser field), then this special mode and the atomic system would be treated as a small quantum system with the remaining modes constituting the reservoir, so that Markovian behavior would still apply for the small system.

In certain cases, however, such as for atoms in high-Q cavities or in photonic band gap materials, either the coupling constants or the mode density (or both) are no longer slowly varying functions, and standard Markovian master equation methods are no longer valid (see Ref. [3] for a recent review). A number of non-Markovian methods have been formulated, see, for example, references in Ref. [4]. These include non-Markovian master equations [5–7], the time-convolutionless projection operator master equation [8], Heisenberg equations of motion [9,10], stochastic wave-function methods for non-Markovian processes [11–16], methods based on the essential states approximation or resolvent operators [3,17–19], the pseudomode approach [20,21], Fano diagonalization Refs. [22] (and [23,4]), and various short-time approximations [24,25]. The last four approaches are easiest to apply, providing clear physical insight into the processes involved.

One such method is that of the pseudomode theory [20,21]. This method was developed for the case of a two-level atom coupled to a structured electromagnetic field reservoir in the vacuum states and was then restricted to single-photon excitations of the reservoir. The treatment started from the time-dependent state vector for the atom-field system, written as a linear combination of one-photon states with the atom in the ground state and vacuum states with the atom in the excited state. The basis of the method was that the atomic dynamics only depended in this case on the behavior of a single function, the reservoir structure function, defined as the product of the mode density and the square of the coupling constant. The complex frequencies and residues of the poles of this function in the lower-half complex frequency plane enabled so-called pseudomodes to be introduced, one for each of the finite number of poles. The non-

Markovian equation for the complex amplitude of the state with the atom excited (and the field in the vacuum state) could then be replaced by Markovian equations involving the *finite* number of pseudomode amplitudes together with the amplitude for the excited atomic state. The pseudomodes are originally related mathematically to the reservoir structure function, but in some cases their physical origin can be explained. For the case of the atom in a high-Q cavity, where the coupling constants vary rapidly near the cavity resonance frequencies while the mode density is slowly varying, the pseudomodes can be interpreted [4] in terms of the cavity quasimodes [26]. For the case of an atom in a photonic band-gap system, no pseudomode theory is yet available, though a treatment in terms of quasimodes [27] can be used to account for the frequency dependence of the coupling constants and mode densities. A treatment of superradiance in a photonic band-gap continuum [28] is based on the idea of replacing the photonic band gap system by a pair of degenerate cavity modes coupled to the multi-atom system and with each other, one of the modes being also coupled to a Markovian bath. In terms of the treatment in Ref. [4], such a case would produce a Fano-profile reservoir structure function, with the Fano window representing the photonic band gap. The two cavity modes would correspond to two pseudomodes. The problem for photonic band-gap situations is that the mode density is actually a discontinuous function of the frequency, and thus the reservoir structure function would not have a finite number of simple poles, though approximate representations of the reservoir structure function in such a form might be found.

Leaving aside the difficulties associated with the pseudomode theory for photonic band-gap systems, it would be desirable to see if pseudomode theory could be extended to cases where multiple-photon excitation of the structured reservoir is involved, as the original treatment [20] only covers single-photon excitation. The limitation of current treatments to the single-photon excitation case has been noted in Ref. [3], but some work has been carried out on cases of multiphoton excitation of the reservoir. Such a multiphoton situation would apply if the two-level atom was replaced by a three-level cascade (or ladder) system, with an initial condition of the atom in the uppermost state and no photons present in the electromagnetic field. The two-step decay will generate electromagnetic field states with two photons present. Another case of multiphoton excitation occurs for an excited two-level atom coupled to a defect mode containing one photon, the atom also being coupled to a photonic band-gap continuum. Beginning with the essential states approximation, a numerical method based on replacing the density of modes by a discretized model has been used in this latter situation [29,30] and in the case of the cascade system [31]. Similar numerical methods have also been used to treat stimulated emission in a photonic crystal [32]. The cascade system case with one transition coupled near resonantly to the edge of a photonic gap (and the other coupled to a flat continuum) has also been treated via the resolvent operator method in Ref. [33]. Although the treatment is analytic, this feature results from being able to ignore processes in which the two emitted photons are produced in a different sequence—a reasonable approximation if the two transition frequencies are very different. However, a more general analytical method would be desirable, and therefore we aim to see if pseudomode theory can be extended to treat the multiphoton excitation case without having to make assumptions about the order in which the photons are produced. Whether an extension is possible involves first showing that the atomic dynamics only depends on the behavior of reservoir structure functions—in a cascade system we would expect there to be more than one reservoir structure function, since two coupling constants are present. A next step would be to then introduce suitable pseudomode amplitudes, based on the poles of the reservoir structure functions and to show that Markovian equations apply to these pseudomode amplitudes.

The present paper shows (following the approach of Ref. [19]) that in the case of a three-level atomic system the dynamics is completely controlled by the reservoir structure functions, and gives several methods for determining the atomic and field behavior. These methods could be applied both to photonic band-gap and high-Q-cavity cases, since the general equations (14)–(23) defining the solution only depend on the reservoir structure functions and not on the specific type of structured reservoir involved. However, as a test, in this paper we only apply the methods to a situation involving a single Lorentzian reservoir structure function. This situation could apply when both cascade transitions have the same frequency and are equally coupled to a single high-Q-cavity mode. We also are able to interpret the results via an equivalent pseudomode model. Situations involving photonic band gaps could be modeled by appropriate reservoir structure functions (see, for example, Refs. [20,28]).

Section II of this paper sets out the theory of non-Markovian dynamical behavior for the three-level cascade system where both transitions are coupled to a single structured reservoir. The state amplitudes are determined from solutions to certain integral equations. Approaches to solving the dynamical equations, including a numerical determination of the excited-state probability for a simple case (and its pseudomode theory interpretation) is presented in Sec. III. Section IV deals with the simpler case of non-Markovian dynamics for the cascade system with the two transitions coupled to two separate reservoirs, again with numerical results presented for comparison to the single reservoir case. An alternative approach to solving the dynamical equations based on nonorthogonal eigenfunction methods is set out in Appendixes A–D. The paper is summarized in Sec. V.

II. DYNAMICAL THEORY FOR A SINGLE RESERVOIR

A. The Hamiltonian

The model system we will consider has a three-level atom, with states denoted $|0\rangle$, $|1\rangle$, and $|2\rangle$, coupled to a reservoir of electromagnetic radiation modes (or heat bath) which is to be at effectively zero temperature. The bath modes will be described by a density ρ_λ, frequency ω_λ, and raising and lowering operators $\hat{a}_\lambda^\dagger$ and $\hat{a}_\lambda$.

The Hamiltonian for the system is given (in the rotating wave approximation) by

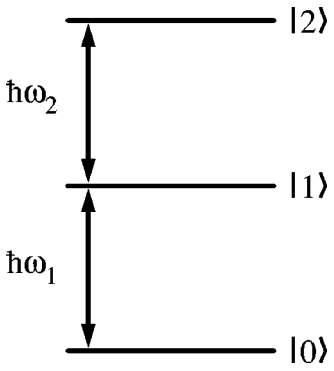

FIG. 1. The three-level cascade (or ladder) atomic system. The atomic states 0, 1, and 2 have transition frequencies ω_1 and ω_2.

$$\hat{H}=\hbar\Big[\omega_1|1\rangle\langle 1|+(\omega_1+\omega_2)|2\rangle\langle 2|+\sum_\lambda \omega_\lambda \hat{a}_\lambda^\dagger \hat{a}_\lambda + \sum_\lambda [g_{\lambda 1}(\hat{a}_\lambda^\dagger|0\rangle\langle 1|+\hat{a}_\lambda|1\rangle\langle 0|) + g_{\lambda 2}(\hat{a}_\lambda^\dagger|1\rangle\langle 2|+\hat{a}_\lambda|2\rangle\langle 1|)]\Big], \quad (1)$$

where the atomic transition frequencies are ω_1 for $0\leftrightarrow 1$ (i.e., between atomic states $|0\rangle$ and $|1\rangle$) and ω_2 for $1\leftrightarrow 2$ (i.e., between atomic states $|1\rangle$ and $|2\rangle$); see Fig. 1. The coupling of electromagnetic radiation with frequency ω_λ to the transition $0\leftrightarrow 1$ involves the frequency-dependent coupling constant $g_{\lambda 1}$. Likewise, $g_{\lambda 2}$ represents the coupling of the electromagnetic radiation field to the $1\leftrightarrow 2$ transition. Real coupling constants will be chosen. It is these frequency-dependent coupling constants combined with the mode density which define the reservoir structure.

B. Coupled amplitude equations

The Schrödinger picture state vector for the three-level cascade atom coupled to the quantum electromagnetic field may be written as

$$|\Psi(t)\rangle=c_2 e^{-i(\omega_1+\omega_2)t}|2\rangle|0_\lambda\rangle+\sum_\lambda c_{1\lambda}e^{-i(\omega_1+\omega_\lambda)t}|1\rangle|1_\lambda\rangle + \sum_\lambda c_{0\lambda\lambda}e^{-2i\omega_\lambda t}|0\rangle|2_\lambda\rangle + \sum_{\lambda,\mu,(\lambda<\mu)} c_{0\lambda\mu}e^{-i(\omega_\lambda+\omega_\mu)t}|0\rangle|1_\lambda 1_\mu\rangle, \quad (2)$$

where $c_2, c_{1\lambda}, c_{0\lambda\lambda}$, and $c_{0\lambda\mu}$ are the amplitudes of the various states in the interaction picture. The radiation states included are as follow: $|0_\lambda\rangle$ in which all the bath modes are in the vacuum state; $|1_\lambda\rangle$ in which the mode with frequency ω_λ has a single excitation, with other modes being in the vacuum state; $|2_\lambda\rangle$ in which the mode with frequency ω_λ has been raised to the second excitation, with other modes being in the vacuum state; and $|1_\lambda 1_\mu\rangle$ in which the modes with frequencies ω_λ and ω_μ have a single excitation, with other modes being in the vacuum state. A convention for an ordered listing of the modes λ for the quantum electromagnetic field is assumed, so that double sum over λ,μ does not lead to a specific state $|1_\lambda 1_\mu\rangle$ being included twice.

The initial-state vector is assumed to be of the form

$$|\Psi(0)\rangle=|2\rangle|0_\lambda\rangle, \quad (3)$$

which allows us to explore the nontrivial case of two photons appearing in the reservoir as a result of the interaction with the excited atom.

Substitution of Eq. (2) into the time-dependent Schrödinger equation gives a closed set of coupled equations for the amplitudes in the situation where the initial state is given by Eq. (3). If we then take Laplace transforms of the the coupled amplitude equations, we obtain the algebraic equations

$$\begin{aligned}
s\bar{c}_2(s)-1&=-i\sum_\lambda g_{\lambda 2}\bar{c}_{1\lambda}(s+i(\omega_\lambda-\omega_2)),\\
s\bar{c}_{1\lambda}(s)&=-i\sum_{\mu,(\mu>\lambda)} g_{\mu 1}\bar{c}_{0\lambda\mu}(s+i(\omega_\mu-\omega_1))\\
&\quad -i\sum_{\mu,(\mu<\lambda)} g_{\mu 1}\bar{c}_{0\mu\lambda}(s+i(\omega_\mu-\omega_1))\\
&\quad -ig_{\lambda 1}\sqrt{2}\bar{c}_{0\lambda\lambda}(s+i(\omega_\lambda-\omega_1))\\
&\quad -ig_{\lambda 2}\bar{c}_2(s+i(\omega_2-\omega_\lambda)),\\
s\bar{c}_{0\lambda\lambda}(s)&=-ig_{\lambda 1}\sqrt{2}\bar{c}_{1\lambda}(s+i(\omega_1-\omega_\lambda)),\\
s\bar{c}_{0\lambda\mu}(s)&=-ig_{\mu 1}\bar{c}_{1\lambda}(s+i(\omega_1-\omega_\mu))\\
&\quad -ig_{\lambda 1}\bar{c}_{1\mu}(s+i(\omega_1-\omega_\lambda)) \quad (\lambda<\mu),
\end{aligned} \quad (4)$$

where the Laplace transforms of the amplitudes are $\bar{c}_2, \bar{c}_{1\lambda}, \bar{c}_{0\lambda\lambda}$ and $\bar{c}_{0\lambda\mu}$ and the transform variable is s. These equations for a cascade system coupled to a single structured reservoir are equivalent to those in Ref. [31]. It should be noted that similar equations are given in Ref. [33] for the case where the two transitions are coupled to two separate reservoirs. This case is treated in Sec. IV. The two separate reservoirs case leads to simpler equations—first, because it is assumed that the λ,μ photons are produced in just one sequence (for example, $|2\rangle|0_\lambda\rangle\rightarrow|1\rangle|1_\lambda\rangle\rightarrow|0\rangle|1_\lambda 1_\mu\rangle$), and second, because states of the form $|0\rangle|2_\lambda\rangle$ are not present. While these may be a good approximation for the single reservoir case when the transition frequencies are very different, the other process ($|2\rangle|0_\lambda\rangle\rightarrow|1\rangle|1_\mu\rangle\rightarrow|0\rangle|1_\lambda 1_\mu\rangle$) would also need to be included when the transition frequencies are similar, such as in a quantum harmonic oscillator or a Rydberg atom.

Following the approach of Ref. [19] we change variables to the reduced amplitudes $\bar{b}_2, \bar{b}_{1\lambda}, \bar{b}_{0\lambda\mu}, \bar{b}_{0\lambda\lambda}$ such that

$$\bar{c}_2=\bar{b}_2,$$

$$\overline{c}_{1\lambda}=g_{\lambda 2}\overline{b}_{1\lambda},$$

$$\overline{c}_{0\lambda\mu}=g_{\lambda 2}g_{\mu 1}\overline{b}_{0\lambda\mu}\quad(\lambda<\mu)$$

$$\overline{c}_{0\lambda\lambda}=g_{\lambda 2}g_{\lambda 1}\overline{b}_{0\lambda\lambda}. \tag{5}$$

Thus we have

$$s\overline{b}_2(s)-1=-i\sum_{\lambda} g_{\lambda 2}^2\overline{b}_{1\lambda}(s+i(\omega_\lambda-\omega_2)), \tag{6}$$

$$\begin{aligned}s\overline{b}_{1\lambda}(s)=&-i\sum_{\mu,(\mu>\lambda)} g_{\mu 1}^2\overline{b}_{0\lambda\mu}(s+i(\omega_\mu-\omega_1))\\&-i\sum_{\mu,(\mu<\lambda)} g_{\mu 1}^2\alpha_{\lambda\mu}\overline{b}_{0\mu\lambda}(s+i(\omega_\mu-\omega_1)),\\&-ig_{\lambda 1}^2\sqrt{2}\overline{b}_{0\lambda\lambda}(s+i(\omega_\lambda-\omega_1))\\&-i\overline{b}_2(s+i(\omega_2-\omega_\lambda)),\end{aligned} \tag{7}$$

$$s\overline{b}_{0\lambda\lambda}(s)=-i\sqrt{2}\overline{b}_{1\lambda}(s+i(\omega_1-\omega_\lambda)), \tag{8}$$

$$\begin{aligned}s\overline{b}_{0\lambda\mu}(s)=&-i\overline{b}_{1\lambda}(s+i(\omega_1-\omega_\mu))\\&-i\alpha_{\lambda\mu}\overline{b}_{1\mu}(s+i(\omega_1-\omega_\lambda))\quad(\lambda<\mu),\end{aligned} \tag{9}$$

where

$$\alpha_{\lambda\mu}=\frac{g_{\lambda 1}g_{\mu 2}}{g_{\lambda 2}g_{\mu 1}}. \tag{10}$$

Equations analogous to Eqs. (6)–(9) are given below in Eqs. (40)–(42) for the case of a cascade system coupled to two separate reservoirs.

C. Reservoir structure functions and integral equation to determine amplitudes

Next we eliminate $\overline{b}_{0\lambda\mu},\overline{b}_{0\lambda\lambda}$ by substitution of Eqs. (8) and (9) in Eq. (7). This gives

$$\begin{aligned}i\overline{b}_2(s)+\sum_{\mu}\Bigg[&\Bigg(s+i(\omega_\lambda-\omega_2)\\&+\sum_{\eta}\frac{g_{\eta 1}^2}{s+i(\omega_\lambda+\omega_\eta-\omega_1-\omega_2)}\Bigg)\delta_{\lambda\mu}\\&+\frac{g_{\mu 1}^2\alpha_{\lambda\mu}}{s+i(\omega_\lambda+\omega_\mu-\omega_1-\omega_2)}\Bigg]\overline{b}_{1\mu}(s+i(\omega_\mu-\omega_2))=0.\end{aligned} \tag{11}$$

Together with Eq. (6) we now have a set of coupled equations for the $\overline{b}_2(s)$ and $\overline{b}_{1\lambda}(s)$. If we eliminate $\overline{b}_2(s)$, we obtain an equation for the $\overline{b}_{1\lambda}$ alone:

$$\begin{aligned}\sum_{\mu}\Bigg[&s\Bigg(s+i(\omega_\lambda-\omega_2)+\sum_{\eta}\frac{g_{\eta 1}^2}{s+i(\omega_\lambda+\omega_\eta-\omega_1-\omega_2)}\Bigg)\delta_{\lambda\mu}\\&+s\frac{g_{\mu 1}^2\alpha_{\lambda\mu}}{s+i(\omega_\lambda+\omega_\mu-\omega_1-\omega_2)}+g_{\mu 2}^2\Bigg]\\&\times\overline{b}_{1\mu}(s+i(\omega_\mu-\omega_2))=-i.\end{aligned} \tag{12}$$

It is useful to rewrite this by dividing by s, using the properties of the Kronecker delta function, and substituting from Eqs. (10) and (12) to obtain

$$\begin{aligned}&\Bigg(s+i(\omega_\lambda-\omega_2)+\sum_{\eta}\frac{g_{\eta 1}^2}{s+i(\omega_\lambda+\omega_\eta-\omega_1-\omega_2)}\Bigg)\overline{b}_{1\lambda}[s\\&\quad+i(\omega_\lambda-\omega_2)]\\&\quad+\sum_{\mu}\Bigg(g_{\mu 1}^2\alpha_{\lambda\mu}\frac{1}{s+i(\omega_\lambda+\omega_\mu-\omega_1-\omega_2)}+\frac{g_{\mu 2}^2}{s}\Bigg)\\&\quad\times\overline{b}_{1\mu}(s+i(\omega_\mu-\omega_2))\\&=\frac{-i}{s}.\end{aligned} \tag{13}$$

We note that the terms involving the quantity $\alpha_{\lambda\mu}$ are absent in similar equations in Ref. [33], resulting in their equations for $\overline{b}_{1\lambda}$ being easily solvable. As mentioned earlier, the additional terms we have result from allowing for photons to be emitted into the single reservoir in two different sequences, an effect not present in the two separate reservoir case treated in Ref. [33]. In our case, we next convert the sums to integrals, i.e., $\Sigma_\mu\rightarrow\int d\omega_\mu\rho(\omega_\mu)$, where $\rho(\omega_\mu)$ is the mode density, so that Eq. (13) can be written in the form of an integral equation

$$A(\omega_\lambda)\overline{f}(\omega_\lambda)+\int d\omega_\mu B(\omega_\lambda,\omega_\mu)\overline{f}(\omega_\mu)=C, \tag{14}$$

with

$$\overline{f}(\omega_\lambda)=\overline{b}_{1\lambda}(s+i(\omega_\lambda-\omega_2)) \tag{15}$$

$$\begin{aligned}A(\omega_\lambda)=&s+i(\omega_\lambda-\omega_2)\\&+\int d\omega_\eta\rho(\omega_\eta)\frac{g_{\eta 1}^2}{s+i(\omega_\lambda+\omega_\eta-\omega_1-\omega_2)},\end{aligned} \tag{16}$$

$$\begin{aligned}B(\omega_\lambda,\omega_\mu)=\rho(\omega_\mu)\Bigg(&g_{\mu 1}^2\frac{g_{\lambda 1}g_{\mu 2}}{g_{\lambda 2}g_{\mu 1}}\frac{1}{s+i(\omega_\lambda+\omega_\mu-\omega_1-\omega_2)}\\&+\frac{g_{\mu 2}^2}{s}\Bigg),\end{aligned} \tag{17}$$

$$C=\frac{-i}{s}. \tag{18}$$

The quantities $\overline{f}(\omega_\lambda)$, $A(\omega_\lambda)$, $B(\omega_\lambda,\omega_\mu)$, and C are, of course, all functions of the Laplace variable s, but for sim-

plicity of notation s is left implicit. The integral equation (14) is a Fredholm integral equation of the second kind (see, e.g., Ref. [34]). Methods of solving such equations include replacing the frequency spaces by grids of points, thereby converting the integral equation into matrix equations that could be solved numerically for each value of s. We will discuss one such approach in Sec. III A. In Appendix D we also discuss a more formal method of solving the integral equation, based on the eigenfunctions of the kernel $B(\omega_\lambda,\omega_\mu)/A(\omega_\lambda)$ and of its adjoint.

We also find it convenient to write the integral equation in the form

$$\overline{f}(\omega_\lambda)+\int d\omega_\mu K(\omega_\lambda,\omega_\mu)\overline{f}(\omega_\mu)=d(\omega_\lambda), \tag{19}$$

where

$$d(\omega_\lambda)=C/A(\omega_\lambda), \tag{20}$$

$$K(\omega_\lambda,\omega_\mu)=B(\omega_\lambda,\omega_\mu)/A(\omega_\lambda). \tag{21}$$

We note that the coupling constants and mode density appear in the integral equation only in the form "ρg^2." These forms are called reservoir structure functions, as there contain all the essential features of the reservoir and its coupling to the atomic system. Specifically, the reservoir structure functions that appear in Eqs. (16) and (17) are

$$R_1(\omega_\lambda)=\rho(\omega_\lambda)g_{\lambda 1}^2,$$

$$R_2(\omega_\lambda)=\rho(\omega_\lambda)g_{\lambda 2}^2. \tag{22}$$

As the coupling constants are proportional to dipole matrix elements multiplied by the square root of the angular frequency, it is clear that the factors $g_{\mu 2}/g_{\mu 1}$ and $g_{\lambda 1}/g_{\lambda 2}$ in Eq. (17) are independent of the frequencies ω_λ and ω_μ. Hence a third reservoir structure function involving the factor $\alpha_{\lambda\mu}$ is not needed. As the dipole matrix elements would essentially cancel out, the factor $\alpha_{\lambda\mu}$ is of order unity.

In principle, we can solve the integral equation and thus determine the $\overline{b}_{1\lambda}[s+i(\omega_\lambda-\omega_2)]$. Furthermore, the solutions obtain their particular form from just the reservoir structure functions, rather than the density of states or coupling constants alone.

Next we see that in the new notation Eq. (6) becomes

$$\overline{b}_2(s)=\frac{1}{s}-\frac{i}{s}\sum_\lambda g_{\lambda 2}^2\overline{b}_{1\lambda}(s+i(\omega_\lambda-\omega_2))$$

$$\equiv\frac{1}{s}-\frac{i}{s}\int d\omega_\lambda\rho(\omega_\lambda)g_{\lambda 2}^2\overline{f}(\omega_\lambda), \tag{23}$$

and again the step to obtaining $\overline{b}_2(s)$ just involves using the reservoir structure function $R_2(\omega_\lambda)$. Note again that $\overline{f}(\omega_\lambda)$ is a function of the Laplace variable s, so the decay of the initial atomic state $|2\rangle$ described by $\overline{b}_2(s)$ is nonexponential in general.

Finally, we note Eqs. (8) and (9) imply that $\overline{b}_{0\lambda\mu}$ and $\overline{b}_{0\lambda\lambda}$ are fully determined once $\overline{b}_2,\overline{b}_{1\lambda}$ are known, and $\alpha_{\lambda\mu}$ [Eq. (10)] introduces no new frequency dependence as it is independent of frequency. Thus all the reduced amplitudes $\overline{b}_2,\overline{b}_{1\lambda},\overline{b}_{0\lambda\mu}$, and $\overline{b}_{0\lambda\lambda}$ can be determined in principle from reservoir structure functions. As we will see next, this is sufficient to determine the reduced density operator describing the atomic system.

Note that the non-Markovian methods could be applied both to photonic band-gap and high-Q-cavity cases, since the general equations (14)–(23) defining the solution only depend on the reservoir structure functions and not on the specific type of structured reservoir involved. Markovian results can be obtained under conditions where the reservoir structure functions $\rho(\omega_\lambda)g_{\lambda 1,\lambda 2}^2$ are slowly varying functions of ω_λ. Certain integrals give a constant term whose imaginary parts are the (formally divergent) frequency shifts and whose real parts are the decay rates for the states $|1\rangle$ and $|2\rangle$.

D. Atomic density operator

The atomic density operator is defined by

$$\hat{\rho}_A=\mathrm{Tr}_F|\Psi\rangle\langle\Psi|, \tag{24}$$

and it is not difficult to show that

$$\hat{\rho}_A=|b_2(t)|^2|2\rangle\langle 2|+\left(\int d\omega_\lambda\rho(\omega_\lambda)g_{\lambda 2}^2|b_{1\lambda}(t)|^2\right)|1\rangle\langle 1|$$

$$+\left(\int\int_{\lambda\leq\mu}d\omega_\lambda d\omega_\mu\rho(\omega_\lambda)\rho(\omega_\mu)\right.$$

$$\left.\times g_{\lambda 2}^2 g_{\mu 1}^2|b_{0\lambda\mu}(t)|^2\right)|0\rangle\langle 0|. \tag{25}$$

Thus we see that the atomic operator only depends on the reduced amplitudes $b_2(t),b_{1\lambda}(t),b_{0\lambda\mu}(t)(\lambda\leq\mu)$ and the reservoir structure functions. As the former can be determined, in principle, from the reservoir structure functions, we see that the behavior of the cascade atom in the structured reservoir is completely determined by the reservoir structure functions [for the initial state given in Eq. (3)].

On the basis of this key result, it would follow that any existing system could be replaced by an equivalent system, provided that the reservoir structure functions were the same in both models. This is the basis of the treatment of superradiance in a photonic band-gap continuum [28], where the photonic band-gap system is replaced by a pair of degenerate cavity modes coupled to the multiple-atom system and with each other, one of the modes being also coupled to a Markovian bath. In terms of the treatment in Ref. [4], such a case would produce the required Fano-profile reservoir structure function, with the Fano window representing the photonic band gap. The two cavity modes would correspond to two pseudomodes.

The absence of any coherence terms in the atomic density operator is a consequence of the choice of initial state, Eq. (3). The choice of a more general initial state (even with no photons present) of the form

$$|\Psi(0)\rangle=(c_2|2\rangle+c_1|1\rangle+c_0|0\rangle)|0_\lambda\rangle \tag{26}$$

would require the introduction of a more general time-dependent state vector $|\Psi(t)\rangle$ than that given in Eq. (2), to include additional states of the form $|0\rangle|0_\lambda\rangle$,$|1\rangle|0_\lambda\rangle$, and $|0\rangle|1_\lambda\rangle$. The amplitudes for these additional states are not coupled to those for the other states included in Eq. (2). Again, the solutions for these amplitudes just involve reservoir structure functions and are analogous to those already discussed in Ref. [20] for the simpler case of a two-level atom coupled to a structured reservoir. However, as indicated above, the atomic density operator would then include coherence terms.

III. SOLUTIONS FOR THE STATE AMPLITUDES

The integral equation (14) can be solved in different ways. These include (a) numerical methods based on converting the integral equation to a matrix equation, (b) expansions using biorthogonal eigenfunctions, and (c) expansions such as the Fredholm expansion [34]. Only the first of these methods will be used here, but as the second approach using biorthogonal eigenfunctions may be used in later work and has not been used previously in quantum optics problems, it is included here in Appendixes A–D for completeness.

A. Numerical solution of the integral equation: Case of Lorentzian reservoir structure function

As an illustration we consider a greatly simplified example of a three-level system coupled to a reservoir with structure. The simplest possible case is that for the same Lorentzian reservoir structure function associated with both transitions, with all the couplings and transition frequencies equal to each other. That is, we choose a single coupling constant g_λ such that

$$g_{\lambda 1}=g_{\lambda 2}=g_\lambda, \tag{27}$$

which amounts to both the dipole moment matrix elements for the transitions being equal. The atom will also have two equally spaced transitions which are resonant with the reservoir structure,

$$\omega_1=\omega_2=\omega_0. \tag{28}$$

We refer only to ω_0 in the following. Thus for the single reservoir structure function we have

$$R_1=R_2=\rho_\lambda g_\lambda^2=\frac{\Gamma\Omega^2}{2\pi}\frac{1}{(\omega_\lambda-\omega_0)^2+(\Gamma/2)^2} \tag{29}$$

as in Ref. [20]. The parameter Ω represents the strength of the coupling and Γ represents the width of the reservoir structure function. This situation would apply for identical cascade transitions coupled to a single high-Q-cavity mode. Cascade transitions in a photonic band-gap reservoir would be treated via a different choice of the reservoir structure functions.

Using this expression for the reservoir structure function we can determine the functions $A(\omega_\lambda)$,$B(\omega_\lambda,\omega_\mu)$, and C in Eqs. (16)–(18) and then the kernel, Eq. (21), becomes

$$K(\omega_\lambda,\omega_\mu)=\frac{\Gamma\Omega^2}{2\pi}\frac{[s+i(\omega_\lambda-\omega_0)+\Gamma/2][2s+i(\omega_\lambda+\omega_\mu-2\omega_0)]}{s[(\omega_\mu-\omega_0)^2+(\Gamma/2)^2][s+i(\omega_\lambda+\omega_\mu-2\omega_0)]Q(\omega_\lambda-\omega_0)}, \tag{30}$$

where $Q(\omega)$ is a quadratic polynomial such that

$$Q(\omega)=(s+i\omega)(s+i\omega+\Gamma/2)+\Omega^2. \tag{31}$$

For this model we thus have an analytic form for the kernel, but to go further it appears that we need to use a numerical method. We could utilize an eigenfunction method, such as that of Appendix D, but choose a very simple approach to solve Eq. (19). The process is simply to represent Eq. (19) as a matrix equation

$$(\mathbf{K}+\mathbf{I})\bar{\mathbf{f}}=\mathbf{d}, \tag{32}$$

where $\mathbf{K}$ and $\mathbf{I}$ are matrices and $\bar{\mathbf{f}}$ and $\mathbf{d}$ are vectors. We then invert $(\mathbf{K}+\mathbf{I})$ to solve for $\bar{\mathbf{f}}$. Thus $K(\omega_\lambda,\omega_\mu)$ is represented at discrete frequency points, in effect a discrete basis of spatial δ functions, e.g., $\mathbf{K}_{\omega_\lambda,\omega_\mu}=K(\omega_\lambda,\omega_\mu)$. Similarly, $\bar{f}(\omega_\lambda)$ and $d(\omega_\lambda)$ are represented at discrete frequency points. From the definition in Eq. (15), we see that if we introduce the function $f(\omega_\lambda,t)$ [which we denote as $f(t)$ for simplicity] via

$$f(\omega_\lambda,t)=\exp[-i(\omega_\lambda-\omega_2)t]b_{1\lambda}(t), \tag{33}$$

then $f(t)$ is the function whose Laplace transform is $\bar{f}(s)\equiv\bar{f}(\omega_\lambda,s)$. However, in order to obtain the real and imaginary parts of $f(t)$, we will need the separate inverse Laplace transforms $\bar{f}_r(s)$,$\bar{f}_i(s)$ which are the Laplace transforms of the real and imaginary parts of $f(t)=f_r(t)+if_i(t)$. For complex s the latter Laplace transforms *cannot* be obtained by

just writing $\overline{f}(s)$ as the sum of its real and imaginary parts. However, the Laplace transform $\overline{f}_r(s)$ of the real part $f_r(t)$ is real [and similarly the Laplace transform $\overline{f}_i(s)$ of the imaginary part $f_i(t)$ is real], if the Laplace transform parameter s is real. Hence, the real and imaginary parts of $\overline{f}(s)$ are equal to the Laplace transforms of the real and imaginary parts of $f(t)$ for s on the real axis, so $\mathrm{Re}\overline{f}(s)=\overline{f}_r(s), \mathrm{Im}\overline{f}(s)=\overline{f}_i(s)$ for s real. As $\overline{f}(s)$ is an analytic function of s, the analytic continuation of $\overline{f}_r(s)+i\overline{f}_i(s)$ from the real axis will determine $\overline{f}(s)$ everywhere.

In this example, if we discretize $K(\omega_\lambda,\omega_\mu)$ on an $N\times N$ grid we define the $N\times N$ complex matrices $\mathbf{K}_r$ and $\mathbf{K}_i$ from the real and imaginary parts of Eq. (30) on the real s axis, and then Eq. (32) becomes

$$\begin{pmatrix} \mathbf{K}_r+\mathbf{I} & -\mathbf{K}_i \\ \mathbf{K}_i & \mathbf{K}_r+\mathbf{I} \end{pmatrix} \begin{pmatrix} \overline{\mathbf{f}}_r \\ \overline{\mathbf{f}}_i \end{pmatrix} = \begin{pmatrix} \mathbf{d}_r \\ \mathbf{d}_i \end{pmatrix}, \tag{34}$$

for s on the real axis. The formal solution for $\overline{\mathbf{f}}_r=\overline{\mathbf{f}}_r(s)$ and $\overline{\mathbf{f}}_i=\overline{\mathbf{f}}_i(s)$ for real s then generates the solution for $\overline{\mathbf{f}}(s)$ everywhere. Because of this [and having first identified $\mathbf{K}_r, \mathbf{K}_i$, $\mathbf{d}_r$ and $\mathbf{d}_i$ for real s using Eqs. (21) and (20)], we can *now* regard Eq. (34) as applying for *all* s. This approach could not be used if the real and imaginary parts of $K(\omega_\lambda,\omega_\mu)$ and $d(\omega_\lambda)$ on the real s axis are not analytic. The matrix inversion step thus involves a matrix with $4N^2$ elements compared to, say, $\mathcal{O}(N^4)$ elements represented by Eqs. (6)–(9) in an equivalent discretized form.

Thus we solve for $\overline{\mathbf{f}}_r$ and $\overline{\mathbf{f}}_i$ in Eq. (34), and hence determine the $\overline{b}_{1\lambda}(s+i(\omega_\lambda-\omega_2))$ of Eq. (15). We then find the $\overline{b}_2(s)$ from the scalar product form Eq. (23) obtained from Eq. (6) so that

$$\overline{b}_{2r}(s)=(1+\mathbf{r}\cdot\overline{\mathbf{f}}_i)/s,$$

$$\overline{b}_{2r}(s)=-\mathbf{r}\cdot\overline{\mathbf{f}}_r/s, \tag{35}$$

where $\mathbf{r}\equiv\{\rho_\lambda g_{\lambda 2}^2\}$. Finally, $b_2(t)$ is determined by a numerical inverse Laplace transform.

Figure 2 shows some results for this numerical matrix approach with the kernel given in Eq. (30), which was derived from the reservoir structure function in Eq. (29). The three curves show the upper-state population for three different sizes of matrix which were used to discretize the integral equation. Each case used the same parameters $\Omega=1$ and $\Gamma=1$, where there is a distinct non-Markovian evolution that could not be treated perturbatively because of the strong coupling to the reservoir structure. The solid curve in Fig. 2 shows a good result that was obtained with a matrix of size 150×150 for this problem. Reducing the matrix size to 100×100 (dashed) results in only a slight degradation of the result. However, further reduction of the matrix size affects the numerical result quite badly.

The effect of changing the coupling strength Ω is shown in Fig. 3. The probability of finding the atomic system in the highest atomic state is shown. For strong coupling [Fig. 3(a)] we see damped oscillations that are a typical manifestation of non-Markovian processes. As the coupling is reduced [Fig. 3(b)], the oscillations weaken and then further reductions in the coupling strength Ω [Fig. 3(c)] result in no oscillations and decay that is closer to exponential and on a longer time scale than the strong coupling cases. Fig. 3(c) still shows some visible initial quadratic behavior because of the relatively high value of Ω/Γ.

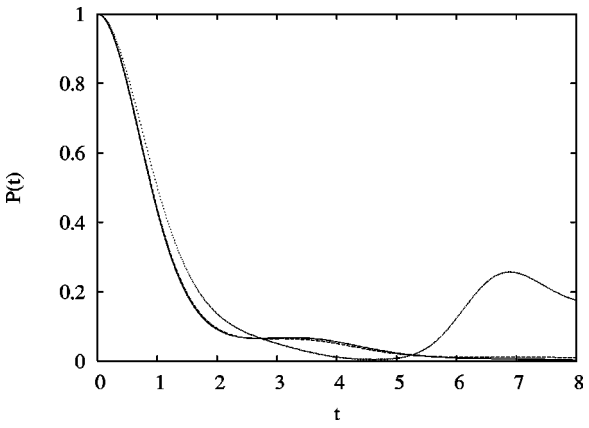

FIG. 2. Time evolution of the probability of finding the system in state 2; $P(t)=|b_2(t)|^2$. The reservoir structure function is given by Eq. (29) with $\Gamma=1$ and $\Omega=1$ in scaled units. The grid size for the discretized kernel was 150×150 (solid), 100×100 (dashed), and 50×50 (dotted). In each case a range of ±30 for $\omega_\lambda-\omega_0$ and $\omega_\mu-\omega_0$ was chosen. The result for a grid size of 150×150 (solid curve) gives a reasonably accurate result.

B. Equivalent pseudo mode model

The reservoir structure function given in Eq. (29) is extremely simple and guided by our previous work we can reproduce the results of Fig. 3, i.e., the population $|b_2(t)|^2$, from the Markovian master equation

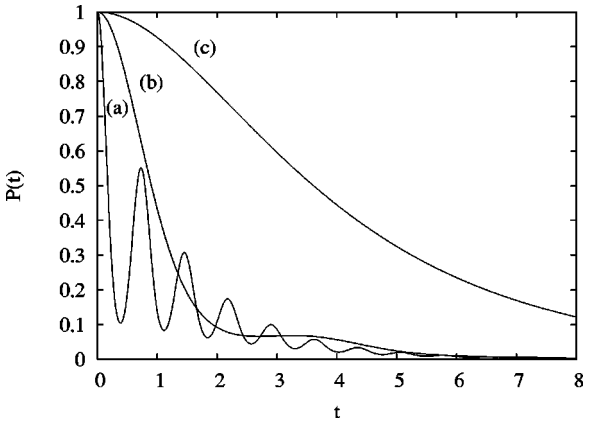

FIG. 3. Time evolution of the probability of finding the system in state 2; $P(t)=|b_2(t)|^2$. The reservoir structure function is Eq. (29) with $\Gamma=1$ and (a) $\Omega=5.0$, (b) $\Omega=1.0$, and (c) $\Omega=0.3$, in scaled units. The grid size for the discretized kernel was 150×150 chosen with a range of ±30 for $\omega_\lambda-\omega_0$ and $\omega_\mu-\omega_0$ in scaled units (as in Fig. 2).

$$\frac{\partial\hat{\rho}}{\partial t}=-i[\hat{V},\hat{\rho}]-\frac{\Gamma}{2}(\hat{a}^{\dagger}\hat{a}\hat{\rho}+\hat{\rho}\hat{a}^{\dagger}\hat{a}\hat{\rho}-2\hat{a}\hat{\rho}\hat{a}^{\dagger}), \quad (36)$$

which is given in the interaction picture with the atom-field coupling term

$$\hat{V}=\Omega(\hat{a}^{\dagger}|0\rangle\langle 1|+\hat{a}|1\rangle\langle 0|+\hat{a}^{\dagger}|1\rangle\langle 2|+\hat{a}|2\rangle\langle 1|). \quad (37)$$

In this master equation we have introduced a single oscillator, or pseudomode [20], which is represented by the harmonic-oscillator operators $\hat{a}$ and $\hat{a}^{\dagger}$. In this approach (see Ref. [20]) pseudomodes are introduced as assumed bosonic entities, rather than via constructing pseudomode amplitudes. A cascade atom resonantly coupled to a damped high-Q-cavity mode, which is also coupled to a Markovian bath of vacuum modes, is an example of a physical system which has the same master equation as Eq. (36). Such a model was considered in our earlier work [4], where we showed that multiple excitations of a structured reservoir could be treated for reservoir structure functions such as Eq. (29). To utilize the present pseudomode model we solve the master equation (36) with the initial condition of an empty pseudomode and the atom in the state $|2\rangle$. On tracing out the pseudomode, to obtain atomic properties alone, we can reproduce the results of the matrix method used with the kernel of Eq. (30). It should be emphasized that it does not appear to be easy to find such a simple master equation for more complex reservoir structures such as photonic band-gap models with branch cuts in the reservoir structure function. In such a case the approach outlined in this paper (which only depends on the reservoir structure functions) may be useful instead. For the present Lorentzian model, the agreement between the matrix method given earlier in this section and the master equation (36) is excellent.

IV. DYNAMICAL THEORY FOR TWO SEPARATE RESERVOIRS

In this paper we have commented in several places that there are differences in our single reservoir treatment from the simpler case of separate reservoirs coupled to the two transitions in our model system. In our model, the two photons may be emitted in either order, whereas with the distinguishable photons in the two reservoir model, only one order of emission is involved. So, with the formalism now complete, it is instructive to look at the explicit differences between our model and the simpler two separate reservoirs model of the kind considered in Ref. [33]. In this case the Hamiltonian in Eq. (1) is replaced by

$$\hat{H}=\hbar\Big[\omega_1|1\rangle\langle 1|+(\omega_1+\omega_2)|2\rangle\langle 2|+\sum_{\lambda}\omega_{\lambda}\hat{a}_{\lambda}^{\dagger}\hat{a}_{\lambda}+\sum_{\mu}\omega_{\mu}\hat{b}_{\mu}^{\dagger}\hat{b}_{\mu}+\sum_{\mu}g_{\mu 1}(\hat{b}_{\mu}^{\dagger}|0\rangle\langle 1|+\hat{b}_{\mu}|1\rangle\langle 0|)+\sum_{\lambda}g_{\lambda 2}(\hat{a}_{\lambda}^{\dagger}|1\rangle\langle 2|+\hat{a}_{\lambda}|2\rangle\langle 1|)\Big], \quad (38)$$

where the bath operators $\hat{a}_{\lambda}^{\dagger}$ and $\hat{a}_{\lambda}$ for the first bath now couple only to the $1\leftrightarrow 2$ transition, and the new bath operators $\hat{b}_{\mu}^{\dagger}$ and $\hat{b}_{\mu}$ for the second bath couple only to the $0\leftrightarrow 1$ transition. For the initial-state vector, Eq. (3), the state vector analogous to Eq. (2) no longer contains a term involving $c_{0\lambda\lambda}$, and there is no restriction over the double sum λ,μ, since the two types of bath modes are now distinct. We can write

$$|\Psi(t)\rangle=c_2e^{-i(\omega_1+\omega_2)t}|2\rangle|0_{\lambda}\rangle|0_{\mu}\rangle+\sum_{\lambda}c_{1\lambda}e^{-i(\omega_1+\omega_{\lambda})t}|1\rangle|1_{\lambda}\rangle|0_{\mu}\rangle+\sum_{\lambda,\mu}c_{0\lambda\mu}e^{-i(\omega_{\lambda}+\omega_{\mu})t}|0\rangle|1_{\lambda}\rangle|1_{\mu}\rangle, \quad (39)$$

involving product states of the atom and one or zero excitation states of the two baths. The equations for the Laplace transforms of the reduced amplitudes, Eqs. (6)–(9), are then replaced by

$$s\overline{b}_2(s)-1=-i\sum_{\lambda}g_{\lambda 2}^2\overline{b}_{1\lambda}(s+i(\omega_{\lambda}-\omega_2)), \quad (40)$$

$$s\overline{b}_{1\lambda}(s)=-i\sum_{\mu}g_{\mu 1}^2\overline{b}_{0\lambda\mu}(s+i(\omega_{\mu}-\omega_1))-i\overline{b}_2(s+i(\omega_2-\omega_{\lambda})), \quad (41)$$

$$s\overline{b}_{0\lambda\mu}(s)=-i\overline{b}_{1\lambda}(s+i(\omega_1-\omega_{\mu})). \quad (42)$$

We note that at this point the differences are that, as well as the absence of the $\overline{b}_{0\lambda\lambda}$ terms, there are no terms involving $\alpha_{\lambda\mu}$ [as in Eqs. (7) and (9)] and there are no restrictions over the sum over μ [as in Eq. (7)] These equations are equivalent to those in Ref. [33].

As in the case of both transitions coupled to one single reservoir, the dynamical behavior only depends on reservoir structure functions, and following the same approach as in Sec. II D it is easy to see that the atomic density operator is also determined from these functions.

If we now follow the elimination procedure of Sec. II C, we find the same equations (14)–(18) for $\overline{f}$, A, B, and C except that the consequence of no $\alpha_{\lambda\mu}$ term being present in Eq. (42) is that the quantity B becomes

$$B(\omega_{\lambda},\omega_{\mu})\rightarrow B(\omega_{\mu})=\rho(\omega_{\mu})\frac{g_{\mu 2}^2}{s}. \quad (43)$$

Crucially B no longer depends on ω_{λ} as previously. Expressions for $\overline{f}$, A, and C are otherwise unchanged.

The integral equation then simplifies to the easily solvable form

$$A(\omega_{\lambda})\overline{f}(\omega_{\lambda})+\int d\omega_{\mu}B(\omega_{\mu})\overline{f}(\omega_{\mu})=C, \quad (44)$$

for which the solution is

$$\overline{f}(\omega_\lambda)=\frac{C}{1+\int d\omega_\mu K(\omega_\mu,\omega_\mu)}\frac{1}{A(\omega_\lambda)}. \tag{45}$$

In this case the equivalent kernel is separable: $K(\omega_\lambda,\omega_\mu)=B(\omega_\mu)/A(\omega_\lambda)$.

We can apply our results to the situation analogous to that treated in Sec. III A, where both reservoirs, though now separate, have identical coupling constants and reservoir structure functions, and the two atomic transitions are equally spaced and resonant with the reservoir structures. We utilize Eqs. (27)–(29) and, for this simple model, the kernel can be easily obtained as

$$K(\omega_\lambda,\omega_\mu)=\frac{\Gamma\Omega^2}{2\pi}\frac{[s+i(\omega_\lambda-\omega_0)+\Gamma/2]}{s[(\omega_\mu-\omega_0)^2+(\Gamma/2)^2]Q(\omega_\lambda-\omega_0)}. \tag{46}$$

This result may be compared to the previous expression in Eq. (30) for the case of a single reservoir.

The integral $\int d\omega_\mu K(\omega_\mu,\omega_\mu)$ can be performed by using a contour in the lower-half plane, and we obtain

$$\int d\omega_\mu K(\omega_\mu,\omega_\mu)=\frac{\Omega^2}{s}\frac{s+\Gamma}{(s+\Gamma/2)(s+\Gamma)+\Omega^2}. \tag{47}$$

We may now find from Eq. (16) that

$$A(\omega_\lambda)=s+i(\omega_\lambda-\omega_0)+\frac{\Omega^2}{s+\Gamma/2+i(\omega_\lambda-\omega_0)} \tag{48}$$

so the solution for $\overline{f}(\omega_\lambda)$ can be obtained from Eq. (45). We find that

$$\overline{f}(\omega_\lambda)=-i\frac{[(s+\Gamma/2)(s+\Gamma)+\Omega^2][s+i(\omega_\lambda-\omega_0)+\Gamma/2]}{(s+\Gamma/2)[s(s+\Gamma)+2\Omega^2]Q(\omega_\lambda-\omega_0)}. \tag{49}$$

A numerical inversion of $\overline{f}(\omega_\lambda)$ can be performed to obtain $b_2(t)$ using the same approach as in Sec. III A. However, the reservoir structure, Eq. (29), is sufficiently simple that a solution for $b_2(t)$ can be found from Eq. (49). We first need to perform the integral in Eq. (23) which is facilitated by the fact that Eq. (49) has no poles in the lower-half complex plane [for $\mathrm{Re}(s)>0$], while the factor $\rho(\omega_\lambda)g_{\lambda 2}^2$ in Eq. (23) has only a single pole in the lower-half complex plane if we use the example given in Eq. (29). Then if we perform the integral of Eq. (23) around the single lower-half plane pole we find that

$$b_2(s)=\frac{1}{s}-\Omega^2\frac{s+\Gamma}{s(s+\Gamma/2)[s(s+\Gamma)+2\Omega^2]}. \tag{50}$$

If we now perform the inverse Laplace transform, we find

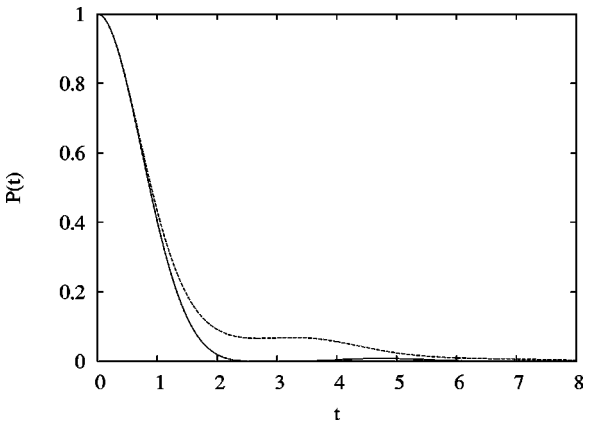

FIG. 4. Time evolution of the probability of finding the system in state 2; $P(t)=|b_2(t)|^2$. The reservoir structure function is Eq. (29) with $\Gamma=1$ and $\Omega=1.0$. The two curves show the effect of changing from two separate reservoirs (solid line) to a single reservoir (dashed line). (Other parameters are as given in Fig. 2. The dashed line in this figure is identical to the solid line in Fig. 2.)

$$b_2(t)=\frac{\Omega^2}{\beta^2}e^{-\Gamma t/2}+\left(1-\frac{\Omega^2}{\beta^2}\right)e^{-\Gamma t/2}\cos(\beta t)+\frac{\Gamma}{2\beta}e^{-\Gamma t/2}\sin(\beta t), \tag{51}$$

where $\beta^2=2\Omega^2-(\Gamma/2)^2$.

The result for the time evolution of the probability for finding the atom in the highest atomic state is seen in Fig. 4. There is clearly a difference from the single reservoir result shown in Fig. 3 (the dashed line in Fig. 4). The present situation, where both atomic transition frequencies are equal and resonant with the structured reservoir, should highlight the difference between the cases of two separate or one single reservoir. In this situation both photons emitted should have similar frequencies, and the single reservoir case where the first emitted photon cannot be distinguished from the other should give different results to the two distinct reservoir case where they can be distinguished.

We note that for strongly coupled systems, $2\Omega^2>(\Gamma/2)^2$, the time evolution in Eq. (51) can be reexpressed in the form

$$b_2(t)=\frac{2\Omega^2}{2\Omega^2-(\Gamma/2)^2}\sin^2(\beta t/2+\phi)e^{-\Gamma t/2}, \tag{52}$$

where

$$\cos\phi=\frac{\Gamma/2}{\sqrt{2}\Omega}. \tag{53}$$

What is interesting here are the oscillations that are given by the square of a sine function, i.e., the probability oscillates as the fourth power of a sine function which is damped at the rate Γ. In the limit $\Omega\gg\Gamma$ the angle ϕ approaches $\pi/2$ and Eq. (52) reduces to $b_2(t)\approx\cos^2(\Omega t/\sqrt{2})e^{-\Gamma t/2}$.

Conversely, for weakly coupled systems, $2\Omega^2 < (\Gamma/2)^2$, the time evolution in Eq. (51) can be reexpressed in the form

$$b_2(t) = \frac{2\Omega^2}{(\Gamma/2)^2 - 2\Omega^2} \sinh^2(\gamma t/2 + \xi) e^{-\Gamma t/2}, \qquad (54)$$

where $\gamma^2 = (\Gamma/2)^2 - 2\Omega^2$ and

$$\cosh \xi = \frac{\Gamma/2}{\sqrt{2}\Omega}. \qquad (55)$$

In the extreme limit of $\Omega \ll \Gamma$, Eq. (54) reduces to the Fermi golden-rule result: $b_2(t) \approx \exp(-2\Omega^2 t/\Gamma)$.

V. CONCLUSION

The dynamical behavior of a three-level atom in a cascade configuration in which both transitions are coupled to a single structured reservoir of electromagnetic field modes, and initially in the upper state, has been analyzed via Laplace transform methods. This situation involves a two-photon excitation of the reservoir, and our equations take into account the two possible sequences in which these two photons are emitted. We have shown that the atomic density operator is determined from the solutions of integral equations, in which the properties of the structured reservoir only appear via so-called reservoir structure functions, all essentially given by the product of the mode density times the square of coupling constants. In the cascade system two distinct reservoir structure functions are involved since there are two transitions. The dependence of the dynamics solely on reservoir structure functions is the necessary condition for treating structured reservoir problems via pseudomode theory, so our results suggest that it may be possible to extend the pseudomode theory to problems involving more than a single-photon excitation of the reservoir.

This result also shows that any existing system could be replaced by an equivalent system, provided that the reservoir structure functions were the same in both models. This is the basis of the treatment of superradiance in a photonic band-gap continuum [28] and the general treatment of multiphoton excitation in terms of quasimodes given in our earlier work [4].

In addition, a similar treatment of the dynamical behavior of a three-level atom in a cascade configuration coupled to two separate structured reservoirs of electromagnetic field modes, and initially in the upper state, has been carried out. One reservoir is coupled to the upper transition, the other to the lower transition. This situation again involves a two-photon excitation of the reservoir, but now only one possible photon emission sequence is involved. In this situation, the equations are simpler, and the integral equation for the amplitudes can be solved analytically. Again, the dynamical features only depend on reservoir structure functions.

A numerical method of solving the integral equations based on discretizing the frequency space has also been obtained, and which can be applied to various structured reservoir situations—such as for high-Q cavities and photonic band-gap systems. Here we have applied this method in a numerical test for a high-Q-cavity situation, where the same Lorentzian reservoir structure function applies to both transitions, showing the non-Markovian decay of the excited state. Results for both the single structured reservoir case and the two separate reservoirs case have been obtained, showing the different behavior in the two cases. This difference is to be expected, as the two photons emitted should have similar frequencies, and only in the two separate reservoirs cases should it be possible to distinguish which order the photons were emitted. In this latter case we were able to solve the model problem analytically. Finally, a formal solution of the integral equations based on the biorthogonal left and right eigenfunctions of the non-Hermitian kernel has been presented for completeness in the appendixes.

Our treatment of the cascade system coupled to a structured reservoir may be compared to those of Ref. [33] in the two separate reservoirs case and to Ref. [31] in the single reservoir case. Both these papers also demonstrate non-Markovian decay of the excited state. Our fundamental amplitude equations in Secs. II B and IV agree with those of these authors. The work in Ref. [31] differs from our treatment, because it is based on replacing the structured reservoir with discrete modes and then using numerical methods. The work in Ref. [33] is analytic. However, a direct comparison of the numerical results is not yet possible with either Ref. [31] or Ref. [33], since both applied their theory to a photonic band-gap system whereas our present application is for the equally important situation of a high-Q cavity. Further applications of our theory involving good analytic approximations to the reservoir structure functions for photonic band-gap systems will, however, enable more detailed comparisons to be made.

ACKNOWLEDGMENTS

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APPENDIX A: INTEGRAL EQUATION KERNEL AND ITS EIGENFUNCTIONS

The kernel $K(\omega_\lambda, \omega_\mu)$ involved in the integral equation (19) and given by Eq. (21) may now be used to define an integral operator $\hat{K}$. The effect of $\hat{K}$ on any function ϕ is defined by

$$(\hat{K}\phi)_{\omega_\lambda} = \int d\omega_\mu K(\omega_\lambda, \omega_\mu) \phi(\omega_\mu). \qquad (A1)$$

The eigenfunctions $\phi_n(\omega_\lambda)$ and eigenvalues ξ_n for the integral operator $\hat{K}$ then satisfy

$$\hat{K}\phi_n = \xi_n \phi_n \qquad (A2)$$

or (in full)

$$\int d\omega_\mu K(\omega_\lambda, \omega_\mu) \phi_n(\omega_\mu) = \xi_n \phi_n(\omega_\lambda). \qquad (A3)$$

Note that we are following Ref. [35] in our definition of the eigenvalue of the integral equation, rather than the definition used in many mathematical textbooks (e.g., Ref. [34]) where $1/\xi_n$ would be the equivalent eigenvalue.

Similarly to Eq. (A1), we can define the adjoint operator $\hat{K}^\dagger$ via

$$(\hat{K}^\dagger\phi)_{\omega_\lambda} = \int d\omega_\mu K^*(\omega_\mu,\omega_\lambda)\phi(\omega_\mu) \tag{A4}$$

with eigenfunctions $\theta_n(\omega_\lambda)$ so that

$$\hat{K}^\dagger\theta_n = \xi_n^*\theta_n . \tag{A5}$$

It is straightforward to show that $\hat{K}^\dagger$ has eigenvalues that are complex conjugates of those for $\hat{K}$ (see Appendix B for details). As $\hat{K}$ will in general be non-Hermitian, the eigenfunctions ϕ_n do not satisfy standard orthogonality conditions. Instead the ϕ_n and the θ_n satisfy so-called biorthogonality conditions

$$\int d\omega_\mu \theta_n^*(\omega_\lambda)\phi_m(\omega_\lambda) = \delta_{nm} . \tag{A6}$$

The normalization result of unity for $n=m$ can be arranged by scaling either the θ_n or ϕ_m by appropriate factors. Although these results are familiar in regard to the mode functions for unstable optical systems ([35,36]), these are not widely used in quantum optics. So, for completeness, a derivation of Eq. (A6) is presented in Appendix C. A formal method of determining the eigenfunctions ϕ_n and θ_n is set out in Appendix B.

APPENDIX B: REPRESENTATION OF THE KERNEL

We expand ϕ_n in an orthonormal basis u_n so that

$$\phi_n(\omega_\lambda) = \sum_m \alpha_m^n u_m(\omega_\lambda) \tag{B1}$$

with

$$\int d\omega_\lambda u_l^*(\omega_\lambda)u_m(\omega_\lambda) = \delta_{lm} . \tag{B2}$$

Then we can write Eq. (A3) as

$$\sum_m \int d\omega_\mu \alpha_m^n K(\omega_\lambda,\omega_\mu)u_m(\omega_\mu) = \xi_n \sum_m \alpha_m^n u_m(\omega_\lambda). \tag{B3}$$

Then if we multiply by $u_l^*(\omega_\lambda)$ and integrate, we find

$$\sum_m \left(\int\int d\omega_\lambda d\omega_\mu u_l^*(\omega_\lambda)K(\omega_\lambda,\omega_\mu)u_m(\omega_\mu) - \delta_{lm}\xi_n \right)\alpha_m^n = 0, \tag{B4}$$

which must be true for all values of l. Equation (B4) is a matrix eigenvalue equation with the matrix

$$K_{lm} = \int\int d\omega_\lambda d\omega_\mu u_l^*(\omega_\lambda)K(\omega_\lambda,\omega_\mu)u_m(\omega_\mu) \tag{B5}$$

and eigenvalues that satisfy

$$|K_{lm} - \xi\delta_{lm}| = 0. \tag{B6}$$

For the operator $\hat{K}^\dagger$ [see Eq. (A4)] the matrix is replaced by its adjoint and clearly its eigenvalues are complex conjugates of those for $\hat{K}$.

The explicit form for K_{lm} is found by substitution of expressions (21) and (17) into Eq. (B5), which yields

$$K_{lm} = \int\int d\omega_\lambda d\omega_\mu u_l^*(\omega_\lambda)\left[\frac{1}{A(\omega_\lambda)} \times \left(\rho(\omega_\mu)g_{\mu 1}^2 \frac{g_{\lambda 1}g_{\mu 2}}{g_{\lambda 2}g_{\mu 1}} \frac{1}{s+i(\omega_\lambda+\omega_\mu-\omega_1-\omega_2)} + \frac{\rho(\omega_\mu)g_{\mu 2}^2}{s} \right) \right] u_m(\omega_\mu). \tag{B7}$$

The integral over ω_μ will involve the reservoir structure functions as defined in Eq. (22). The function $A(\omega_\lambda)$ is also obtainable from the reservoir structure functions [see Eq. (16)]. Note that $g_{\lambda 1}g_{\mu 2}/g_{\lambda 2}g_{\mu 1}$ is independent of frequency in Eq. (B7).

In summary, the matrix K_{lm} and hence the eigenfunctions ϕ_n, θ_n and eigenvalues are all obtained from the reservoir structure functions and given functions, such as the basis set u_n.

APPENDIX C: BIORTHOGONALITY OF EIGENFUNCTIONS

To show that the eigenfunctions satisfy a biorthogonality condition we first write from Eqs. (A2) and (A3), (A4), (A5)

$$\int d\omega_\mu K(\omega_\lambda,\omega_\mu)\phi_n(\omega_\mu) = \xi_n\phi_n(\omega_\lambda),$$

$$\int d\omega_\mu K(\omega_\mu,\omega_\lambda)\theta_m^*(\omega_\mu) = \xi_m\theta_m^*(\omega_\lambda). \tag{C1}$$

After multiplying the first equation by $\theta_m^*(\omega_\lambda)$, the second by $\phi_n(\omega_\lambda)$, and then integrating over ω_λ, we find that

$$\int\int d\omega_\lambda d\omega_\mu \theta_m^*(\omega_\lambda)K(\omega_\lambda,\omega_\mu)\phi_n(\omega_\mu) = \xi_n\int d\omega_\lambda \theta_m^*(\omega_\lambda)\phi_n(\omega_\lambda),$$

$$\int\int d\omega_\lambda d\omega_\mu \phi_n(\omega_\lambda)K(\omega_\lambda,\omega_\mu)\theta_m^*(\omega_\mu) = \xi_m\int d\omega_\lambda \theta_m^*(\omega_\lambda)\phi_n(\omega_\lambda). \tag{C2}$$

After a change of variable in the second equation, the left-hand sides are equal and we then conclude that

$$(\xi_n - \xi_m) \int d\omega_\lambda \theta_m^*(\omega_\lambda) \phi_n(\omega_\lambda) = 0, \tag{C3}$$

so that the biorthogonality condition

$$\int d\omega_\lambda \theta_m^*(\omega_\lambda) \phi_n(\omega_\lambda) = 0 \tag{C4}$$

applies unless $\xi_m = \xi_n$.

APPENDIX D: INTEGRAL EQUATION SOLUTION IN TERMS OF EIGENFUNCTIONS OF *K*

We will *assume* that the set of eigenfunctions ϕ_n form a basis for expanding the solution $\overline{f}(\omega_\lambda)$ [to Eq. (19)]. Likewise we will assume that $d(\omega_\lambda)$ can be expanded in terms of the ϕ_n so that

$$\overline{f}(\omega_\lambda) = \sum_n \overline{f}_n \phi_n(\omega_\lambda),$$

$$d(\omega_\lambda) = \sum_n d_n \phi_n(\omega_\lambda). \tag{D1}$$

Using the biorthogonality of the eigenfunctions [Eq. (A6)] the expansion coefficients can be found as

$$\overline{f}_n = \int d\omega_\lambda \theta_n^*(\omega_\lambda) \overline{f}(\omega_\lambda),$$

$$d_n = \int d\omega_\lambda \theta_n^*(\omega_\lambda) d(\omega_\lambda). \tag{D2}$$

Substituting Eq. (D1) into Eq. (19) and using the eigenvalue equation (A3) we find that

$$\sum_n \overline{f}_n \phi_n(\omega_\lambda) + \sum_n \overline{f}_n \int d\omega_\mu K(\omega_\lambda, \omega_\mu) \phi_n(\omega_\mu)$$

$$= \sum_n d_n \phi_n(\omega_\lambda),$$

$$\sum_n (\overline{f}_n + \xi_n \overline{f}_n - d_n) \phi_n(\omega_\lambda) = 0. \tag{D3}$$

Using the biorthogonality result for the eigenvalue ξ_n we see that

$$\overline{f}_n (1 + \xi_n) - d_n = 0, \tag{D4}$$

so that provided $\xi_n \neq -1$

$$\overline{f}_n = \frac{d_n}{1 + \xi_n}, \tag{D5}$$

which gives the solutions for the expansion coefficients for $\overline{f}(\omega_\lambda)$ in terms of known quantities. The quantities $\overline{f}_n, \phi_n(\omega_\lambda), K(\omega_\lambda, \omega_\mu)$, and ξ_n are, of course, all functions of the Laplace variable *s*, but for simplicity of notation *s* is left implicit.

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